

#### PATENT APPLICATION

#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

Hirohisa Tanaka et al.

Appln. No.: 10/520,068

Group Art Unit: 1754

Filed: January 5, 2005

Examiner: Timothy C. VANOY

For: METHOD FOR PRODUCING PEROVSKITE-TYPE COMPOSITE OXIDE

## DECLARATION UNDER 37 C.F.R. § 1.132

United States Patent and Trademark Office Commissioner for Patents P.O. Box 1450 Alexandria, Virginia 22313-1450

Sir:

I, Kimiyoshi KANEKO, hereby declare and state:

THAT I am a citizen of Japan;

THAT I received a Degree from the Division of Chemistry in the Faculty of Science of Hokkaido University in March, 1969;

THAT I was employed by Hokko Chemical Industry Co., Ltd. from April, 1969, ultimately holding the position of Group Leader of the Fine Ceramics in the Fine Chemicals Development Division, and that I engaged in the development and production of the METHOD FOR PRODUCING PEROVSKITE-TYPE COMPOSITE OXIDE claimed in the present application;

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THAT I became a co-inventor of the above-identified application;

THAT I have reviewed the Office Actions of June 5, 2007;

{DC001278;1}

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**EXAMPLE 1** 

Preparation of Sr<sub>3</sub>NiPtO<sub>6</sub> (Pt content: 31.84 % by weight)

After toluene was distilled off from 55.34 g of toluene solution of strontium isopropoxide

(0.0492 mol), a mixed solution containing 213 g of 2-methoxyethanol and 640 g of isopropyl

alcohol was added thereto for dissolution. Subsequently, 6.450 g (0.0164 mol) of platinum

acetylacetonate was added and the solution was heated at 70°C for three hours.

Then, 4.081 g (0.0164 mol) of nickel acetate tetrahydrate was added and the solution was

heated at 70°C for one hour. After the heating, 1.77 g of deionized water was added and the

solution was heated at 70°C for two hours.

Thereafter, the resulting mixture was separated from the solvent by distillation, dried in a

vacuum at 100°C, and baked at 500°C for three hours or 1000°C for one hour to prepare Sr<sub>3</sub>NiPtO<sub>6</sub>.

EXAMPLE 2

Preparation of Sr<sub>4</sub>PtO<sub>6</sub> (Pt content: 30.41 % by weight)

After toluene was distilled off from 71.99 g of toluene solution of strontium isopropoxide

(0.0640 mol), a mixed solution containing 208 g of 2-methoxyethanol and 624 g of isopropyl

alcohol was added thereto for dissolution. Subsequently, 6.293 g (0.0160 mol) of platinum

acetylacetonate was added and the solution was heated at 70°C for three hours.

Then, 1.77 g of deionized water was added and the solution was heated at 70°C for two

hours.

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Thereafter, the resulting mixture was separated from the solvent by distillation, dried in a

vacuum at 100°C, and baked at 500°C for three hours or 1000°C for one hour to prepare Sr<sub>4</sub>PtO<sub>6</sub>.

EXAMPLE 3

Preparation of MgAl<sub>2</sub>O<sub>4</sub> Supporting Sr<sub>3</sub>NiPtO<sub>6</sub> (Pt content: 1.25 % by weight)

After toluene was distilled off from 0.562 g of toluene solution of strontium isopropoxide

(0.5 mmol), a mixed solution containing 11 g of 2-methoxyethanol and 33 g of isopropyl alcohol

was added thereto for dissolution. Subsequently, 0.0657 g (0.167 mmol) of platinum

acetylacetonate was added and the solution was heated at 70°C for three hours.

The obtained solution was added dropwise to a suspension prepared by mixing 2.50 g of

MgAl<sub>2</sub>O<sub>4</sub> (specific surface area of 37 m<sup>2</sup>/g), 11 g of 2-methoxyethanol and 33 g of isopropyl

alcohol, and the suspension was heated at 70°C for one hour. Then, 0.0415 g (0.167 mmol) of

nickel acetate tetrahydrate was added thereto and the suspension was heated at 70°C for one hour.

After the heating, 0.018 g of deionized water was added and the suspension was heated at 70°C for

two hours.

Thereafter, the resulting mixture was separated from the solvent by distillation, dried in a

vacuum at 100°C, and baked at 500°C for three hours or 1000°C for one hour to prepare MgAl<sub>2</sub>O<sub>4</sub>

supporting Sr<sub>3</sub>NiPtO<sub>6</sub>.

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**EXAMPLE 4** 

Preparation of MgAl<sub>2</sub>O<sub>4</sub> Supporting Sr<sub>4</sub>PtO<sub>6</sub> (Pt content: 1.25 % by weight)

After toluene was distilled off from 0.751 g of toluene solution of strontium isopropoxide

(0.668 mmol), a mixed solution containing 11 g of 2-methoxyethanol and 33 g of isopropyl alcohol

was added thereto for dissolution. Subsequently, 0.0657 g (0.167 mmol) of platinum

acetylacetonate was added and the solution was heated at 70°C for three hours.

The obtained solution was added dropwise to a suspension prepared by mixing 2,50 g of

MgAl<sub>2</sub>O<sub>4</sub> (specific surface area of 37 m<sup>2</sup>/g), 11 g of 2-methoxyethanol and 33 g of isopropyl

alcohol, and the suspension was heated at 70°C for one hour. Then, 0.024 g of deionized water was

added and the suspension was heated at 70°C for two hours.

Thereafter, the resulting mixture was separated from the solvent by distillation, dried in a

vacuum at 100°C, and baked at 500°C for three hours or 1000°C for one hour to prepare MgAl<sub>2</sub>O<sub>4</sub>

supporting Sr<sub>4</sub>PtO<sub>6</sub>.

**EVALUATION** 

Reduction/Oxidation Treatment

Each powder of Sr<sub>3</sub>NiPtO<sub>6</sub>, Sr<sub>4</sub>PtO<sub>6</sub>, MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>3</sub>NiPtO<sub>6</sub> and MgAl<sub>2</sub>O<sub>4</sub>

supporting Sr<sub>4</sub>PtO<sub>6</sub> obtained above was subjected to a reduction treatment under a reductive

atmosphere (CO: 7.5 %, H2: 2.5 %, CO2: 8.0 %, and N2: Balanced) at 800°C for 1 hour, and

subsequently to an oxidation treatment under an atmospheric atmosphere at 800°C for one hour.

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#### X-Ray Diffraction Measurement

Each powder mentioned above was measured by X-ray diffraction. The results are shown below:

## 1) Sr<sub>3</sub>NiPtO<sub>6</sub> (Baking Temperature: 500°C)

After the bake, no crystal structure of Sr<sub>3</sub>NiPtO<sub>6</sub> was detected. More specifically. Sr<sub>3</sub>NiPtO<sub>6</sub> was not produced through the baking at 500°C.

#### 2) Sr<sub>3</sub>NiPtO<sub>6</sub> (Baking Temperature: 1000°C)

After the bake, a crystal structure of Sr<sub>3</sub>NiPtO<sub>6</sub> was detected. However, no crystal structure of Sr<sub>3</sub>NiPtO<sub>6</sub> was detected after the reduction treatment and the oxidation treatment. More specifically, it seemed that the Sr<sub>3</sub>NiPtO<sub>6</sub> was disintegrated after the reduction treatment and the oxidation treatment.

# 3) Sr<sub>4</sub>PtO<sub>6</sub> (Baking Temperature: 500°C)

After the bake, no crystal structure of Sr<sub>4</sub>PtO<sub>6</sub> was detected. More specifically, Sr<sub>4</sub>PtO<sub>6</sub> was not produced through the baking at 500°C.

### 4) Sr<sub>4</sub>PtO<sub>6</sub> (Baking Temperature: 1000°C)

After the bake, a crystal structure of Sr<sub>4</sub>PtO<sub>6</sub> was detected. However, no crystal structure of Sr<sub>4</sub>PtO<sub>6</sub> was detected after the reduction treatment and the oxidation treatment. More

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specifically, it seemed that the Sr<sub>4</sub>PtO<sub>6</sub> was disintegrated after the reduction treatment and the oxidation treatment.

5) MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>3</sub>NiPtO<sub>6</sub> (Baking Temperature: 500°C)

After the bake, no crystal structure of Sr<sub>3</sub>NiPtO<sub>6</sub> in MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>3</sub>NiPtO<sub>6</sub> was detected. More specifically, the Sr<sub>3</sub>NiPtO<sub>6</sub> was not produced in MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>3</sub>NiPtO<sub>6</sub> through the baking at 500°C.

6) MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>3</sub>NiPtO<sub>6</sub> (Baking Temperature: 1000°C)

After the bake, no crystal structure of Sr<sub>3</sub>NiPtO<sub>6</sub> in MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>3</sub>NiPtO<sub>6</sub> was detected. More specifically, Sr<sub>3</sub>NiPtO<sub>6</sub> was not produced in MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>3</sub>NiPtO<sub>6</sub> through the baking even at 1000°C.

7) MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>4</sub>PtO<sub>6</sub> (Baking Temperature: 500°C)

After the bake, no crystal structure of Sr<sub>4</sub>PtO<sub>6</sub> in MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>4</sub>PtO<sub>6</sub> was detected. More specifically, the Sr<sub>4</sub>PtO<sub>6</sub> was not produced in MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>4</sub>PtO<sub>6</sub> through the baking at 500°C.

8) MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>4</sub>PtO<sub>6</sub> (Baking Temperature: 1000°C)

After the bake, no crystal structure of Sr<sub>4</sub>PtO<sub>6</sub> in MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>4</sub>PtO<sub>6</sub> was detected. More specifically, the Sr<sub>4</sub>PtO<sub>6</sub> was not produced in MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>4</sub>PtO<sub>6</sub> through the baking even at 1000°C.

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I, the undersigned, declare that all statement made herein on my knowledge are true and that

all statements made on information and belief are believed to be true: and further that these

statements were made with the knowledge that willful false statements and the like so made are

punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States

Code and such willful false statements may jeopardize the validity of the application or any issuing

thereon.

Signed this

day of October, 2007.

19. October 2007.

Kimiyoshi KANEKO

Kimiyoski Kaneki